526 Rec'd PCT/PTO 12 JUL 2000

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		Attorney's Docket Number 05725.0623 U.S. Application No. 09/600132 PCT
International Application. No.	International Filing Date	Priority Date Claimed
PCT/FR98/02805	December 21, 1998	January 13, 1998

Title of Invention:
DYEING COMPOSITION CONTAINING A LACCASE AND KERATINOUS FIBRE DYEING METHOD

Applicant(s) For DO/EO/U-S:
Gérard LANG and Jean COTTERET

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. [X] This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.
2. [ ] This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.
3. [ ] This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. [X] A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. [X] A copy of the International Application as filed (35 U.S.C. 371(c)(2))
   a. [ ] is transmitted herewith (required only if not transmitted by the International Bureau).
   b. [X] has been transmitted by the International Bureau.
   c. [ ] is not required, as the application was filed in the United States Receiving Office (RO/US).
6. [X] A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. [X] Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)).
   a. [ ] are transmitted herewith (required only if not transmitted by the International Bureau).
   b. [ ] have been transmitted by the International Bureau.
   c. [ ] have not been made; however, the time limit for making such amendments has NOT expired.
   d. [X] have not been made and will not be made.
8. [ ] A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. [ ] An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. [ ] A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern other document(s) or information included:

11. [ ] An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. [ ] An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. [ ] A FIRST preliminary amendment.
    [ ] A SECOND or SUBSEQUENT preliminary amendment.
14. [ ] A substitute specification.
15. [ ] A change of power of attorney and/or address letter.

16. [ ] Other items or information:
    a. [ ] Verified Small Entity Statement.
    b. [ ] Copy of Notification of Missing Requirements.

U.S. APPLICATION NO.	INTERNATIONAL APPLICATION NO.	ATTORNEY DOCKET NUMBER
09/600132	PCT/FR98/02805	534 Rec'd PCT/PTO 12 JUL 2000

17. [X] The following fees are submitted: | CALCULATIONS

Basic National Fee (37 CFR 1.492(a)(1)-(5)):

Search Report has been prepared by the EPO or JPO.....$840.00

International preliminary examination fee paid to USPTO (37 CFR 1.482).....$670.00

No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)).....$690.00

Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO.....$970.00

International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4).....$ 96.00

ENTER APPROPRIATE BASIC FEE AMOUNT = $ 840.00

Surcharge of $130.00 for furnishing the oath or declaration later than [ ] 20 [ ] 30 months from the earliest claimed priority date (37 CFR 1.492(e)). $

Claims	Number Filed	Number Extra	Rate	
Total Claims	24 -20=	4	X $18.00	$ 72.00
Independent Claims	1 - 3=		X $78.00	$
Multiple dependent claim(s) (if applicable)			+$260.00	$ 260.00

TOTAL OF ABOVE CALCULATIONS = $1,172.00

Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28) $

SUBTOTAL = $1,172.00

Processing fee of $130.00 for furnishing the English translation later than [ ] 20 [ ] 30 months from the earliest claimed priority date (37 CFR 1.492(f)). + $

TOTAL NATIONAL FEE = $1,172.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). $40.00 per property + $

TOTAL FEES ENCLOSED = $1,172.00

Amount to be refunded $

charged $

a. [X] A check in the amount of **$1,172.00** to cover the above fees is enclosed.

b. [ ] Please charge my Deposit Account No. _____ in the amount of $_____ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. [X] The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 06-0916. A duplicate copy of this sheet is enclosed.

The Commissioner is hereby authorized to charge any other fees due under 37 C.F.R. §1.16 or §1.17 during the pendency of this application to our Deposit Account No. 06-0916.

SEND ALL CORRESPONDENCE TO:
Finnegan, Henderson, Farabow
Garrett & Dunner, L.L.P.
1300 I Street, N.W.
Washington, D.C. 20005-3315
EFC/FPD/rgm

Ernest F. Chapman
Reg. No. 25,961

Submitted: July 12, 2000

PATENT

Attorney Docket No. 05725.0623-00

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re National Stage of International Application No.: PCT/FR98/02805 of:	
Gérard LANG, et al.	
Serial No.: 09/600132	Group Art Unit: Unassigned
PCT Filed: December 21, 1998	Examiner: Unassigned
National Stage Entry: July 12, 2000	
For: DYEING COMPOSITION CONTAINING A LACCASE AND KERATINOUS FIBRE DYEING METHOD	

PRELIMINARY AMENDMENT

BOX PCT
Assistant Commissioner for Patents
Washington, D.C. 20231

Sir:

Prior to the examination of the above-identified application, please amend this application as follows:

IN THE SPECIFICATION:

Please amend the specification as follows:

Page 8, line 7, please replace "$2x10^6$ lacu" with --$2x10^6$ ulac--.

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IN THE CLAIMS:

Please cancel claims 1-22 and add new claims 23-62 as follows:

--23. A composition for the oxidation dyeing of keratinous fibers comprising:

(a) at least one enzyme of the laccase type;

(b) at least one alkalinizing agent chosen from:

(i) basic amino acids;

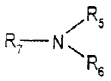
(ii) compounds of the following formula (A):

$$X(OH)_n$$

wherein n is equal to 1 or 2; X is chosen from K, Li and $N^+R_1R_2R_3R_4$ wherein R_1, R_2, R_3, and R_4, which are identical or different, are each chosen from C_1-C_4 alkyl groups, C_1-C_4 monohydroxyalkyl groups and C_2-C_4 polyhydroxyalkyl groups when n=1; or

X is chosen from Mg and Ca when n=2;

(iii) compounds of the following formula (B):



wherein R_5 is chosen from hydrogen groups, C_1-C_6 alkyl groups, C_1-C_6 monohydroxyalkyl groups, and C_2-C_6 polyhydroxyalkyl groups;

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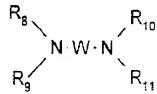
R_6 and R_7, which are identical or different, are each chosen from hydrogen groups, C_1-C_6 alkyl groups, C_1-C_6 monohydroxyalkyl groups, and C_2-C_6 polyhydroxyalkyl groups;

with the proviso that R_5, R_6, and R_7 are not simultaneously chosen from C_2 β-hydroxyalkyl groups;

with the additional proviso that if R_6 and R_7 are simultaneously chosen from H, then R_5 is not chosen from C_2 monohydroxyalkyl groups and branched C_4 monohydroxyalkyl groups;

and with the additional proviso that if R_5 is chosen from H and C_1-C_6 alkyl groups and simultaneously R_6 is chosen from C_1-C_6 alkyl groups, then R_7 is not chosen from H and C_1-C_6 alkyl groups; and

(iv) compounds of the following formula (C):



(C)

wherein W is chosen from propylene groups optionally substituted with a substituent chosen from hydroxyl groups and C_1-C_4 alkyl groups; R_8, R_9, R_{10} and R_{11}, which are identical or different, are each chosen from hydrogen groups, C_1-C_4 alkyl groups and C_1-C_4 hydroxyalkyl groups; and

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(c) at least one oxidation dye with the proviso that said at least one oxidation dye is not chosen from autooxidizable indole dyes.

24. A composition according to Claim 23, wherein said at least one enzyme of the laccase type is chosen from laccases of plant origin, animal origin, fungal origin, and bacterial origin and laccases obtained by biotechnology.

25. A composition according to Claim 23, wherein said at least one enzyme of the laccase type is chosen from those produced by plants performing chlorophyll synthesis.

26. A composition according to Claim 23, wherein said at least one enzyme of the laccase type is chosen from those extracted from plants chosen from Anacardiaceae, Podocarpaceae, Rosmarinus off., Solanum tuberosum, Iris sp., Coffea sp., Daucus carrota, Vinca minor, Persea americana, Catharenthus roseus, Musa sp., Malus pumila, Gingko biloba, Monotropa hypopithys, Aesculus sp., Acer pseudoplatanus, Prunus persica and Pistacia palaestina.

27. A composition according to Claim 23, wherein said at least one enzyme of the laccase type is chosen from those derived from fungi chosen form Pyricularia orizae, Polyporus versicolor, Rhizoctonia praticola, Rhus vernicifera, Scytalidium, Polyporus pinsitus, Myceliophtora thermophila, Rhizoctonia solani, Tramates versicolor, Fomes fomentarius, Chaetomium thermophile, Neurospora crassa, Coriolus versicol, Botrytis cinerea, Rigidoporus lignosus, Phellinus noxius, Pleurotus ostreatus, Aspergillus nidulans, Podospora anserina, Agaricus bisporus, Ganoderma lucidum, Glomerella cingulata, Lactarius piperatus, Russula delica,

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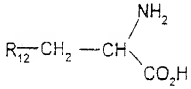
Heterobasidion annosum, Thelephora terrestris, Cladosporium cladosporioides, Cerrena unicolor, Coriolus hirsutus, Ceriporiopsis subvermispora, Coprinus cinereus, Panaeolus papilionaceus, Panaeolus sphinctrinus, Schizophyllum commune, Dichomitius squalens and variants of all said fungi.

28. A composition according to Claim 23, wherein said at least one enzyme of the laccase type is present in a quantity ranging from 0.5 to 2000 lacu units per 100 g of said composition.

29. A composition according to Claim 23, wherein said at least one enzyme of the laccase type is present in a quantity ranging from 1000 to $4x10^7$ u units per 100 g of said composition.

30. A composition according to Claim 23, wherein said at least one enzyme of the laccase type is present in a quantity ranging from 20 to $2x10^6$ ulac units per 100 g of said composition.

31. A composition according to Claim 23, wherein said basic amino acids are chosen from the following formula (D):



(D)

wherein R_{12} is chosen from:

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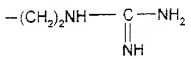
;

$-(CH_2)_3NH_2$;

$-(CH_2)_2NH_2$;

$-(CH_2)_2NHCONH_2$;



;

32. A composition according to Claim 23, wherein said compounds of formula (B) are chosen from diethanolamine, monoisopropanolamine, diisopropanolamine, triisopropanolamine, 2-amino-2-methyl-1,3-propanediol,

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2-amino-2-ethyl-1,3-propanediol, 2-amino-1-n-butanol, 1-diethylamino-2,3-propanediol, tris(hydroxymethyl)aminomethane and ethylmonoethanolamine.

33. A composition according to Claim 23, wherein said at least one alkalinizing agent is present in a quantity ranging from 0.001% to 20% by weight relative to the total weight of said composition.

34. A composition according to Claim 33, wherein said at least one alkalinizing agent is present in a quantity ranging from 0.01% to 5% by weight relative to the total weight of said composition.

35. A composition according to Claim 34, wherein said at least one alkalinizing agent is present in a quantity ranging from 0.05% to 3% by weight relative to the total weight of said composition.

36. A composition according to Claim 23, wherein said at least one oxidation dye is at least one oxidation base chosen from ortho- and para-phenylenediamines, ortho- and para-aminophenols, heterocyclic bases, and the acid addition salts of all said oxidation bases.

37. A composition according to Claim 36, wherein said at least one oxidation base is present in a concentration ranging from 0.0005% to 12% by weight relative to the total weight of said composition.

38. A composition according to Claim 37, wherein said at least one oxidation base is present in a concentration ranging from 0.005% to 6% by weight relative to the total weight of said composition.

39. A composition according to Claim 36, wherein said acid

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addition salts are chosen from hydrochlorides, hydrobromides, sulphates, tartrates, lactates and acetates.

40. A composition according to Claim 23, wherein said at least one oxidation dye is at least one coupler chosen from meta-phenylenediamines, meta-aminophenols, meta-diphenols, heterocyclic couplers and the acid addition salts of all said couplers.

41. A composition according to Claim 40, wherein said at least one coupler is chosen from 2-methyl-5-aminophenol, 5-N-(β-hydroxyethyl)amino-2-methylphenol, 3-aminophenol, 1,3-dihydroxybenzene, 1,3-dihydroxy-2-methylbenzene, 4-chloro-1,3-dihydroxybenzene, 2,4-diamino-1-(β-hydroxyethyloxy)benzene, 2-amino-4-(β-hydroxyethylamino)-1-methoxybenzene, 1,3-diaminobenzene, 1,3-bis(2,4-diaminophenoxy)propane, sesamol, α-naphthol, 6-hydroxyindole, 4-hydroxyindole, 4-hydroxy-N-methylindole, 6-hydroxyindoline, 2,6-dihydroxy-4-methylpyridine, 1-H-3-methylpyrazol-5-one, 1-phenyl-3-methylpyrazol-5-one, 2,6-dimethylpyrazolo[1,5-b]-1,2,4-triazole, 2,6-dimethyl[3,2-c]-1,2,4-triazole, 6-methylpyrazolo[1,5-a]benzimidazole and the acid addition salts of all said couplers.

42. A composition according to Claim 40, wherein said at least one coupler is present in a concentration ranging from 0.0001% to 10% by weight relative to the total weight of said composition.

43. A composition according to Claim 42, wherein said at least one

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coupler is present in a concentration ranging from 0.005% to 5% by weight relative to the total weight of said composition.

44. A composition according to Claim 40, wherein said acid salts are chosen from hydrochlorides, hydrobromides, sulphates, tartrates, lactates and acetates.

45. A composition according to Claim 23, further comprising at least one direct dye.

46. A composition according to Claim 45, wherein said at least one direct dye is chosen from nitro, azo and anthraquinone dyes.

47. A composition according to Claim 23, further comprising at least one carrier appropriate for keratinous fibers.

48. A composition according to Claim 47, wherein said at least one carrier is chosen from water and at least one organic solvent.

49. A composition according to Claim 48, wherein said at least one organic solvent is present in a concentration ranging from 1% to 40% by weight relative to the total weight of said composition.

50. A composition according to Claim 49, wherein said at least one organic solvent is present in a concentration ranging from 5% to 30% by weight relative to the total weight of said composition.

51. A composition according to Claim 23, having a pH from about 4 to about 11.

52. A composition according to Claim 51, wherein said pH varies

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from about 6 to about 9.

53. A composition according to Claim 23, further comprising at least one suitable cosmetic adjuvant chosen from surfactants, polymers, thickeners, antioxidants, enzymes different from said at least one enzyme of the laccase type as defined in Claim 23, penetrating agents, sequestering agents, perfumes, dispersing agents, film-forming agents, screening agents, vitamins, preservatives and opacifying agents.

54. A composition according to Claim 23 in the form of an aqueous or aqueous/alcoholic lotion, a gel, a milk, a cream, an emulsion, a thickened lotion or a thickened foam.

55. A composition according to Claim 23, wherein said composition is ready-to-use.

56. A composition according to Claim 23, wherein said keratinous fibers are human keratinous fibers.

57. A composition according to Claim 56, wherein said human keratinous fibers are hair.

58. A method of dyeing keratinous fibers comprising applying to said keratinous fibers for a sufficient time to develop a desired color at least one dyeing composition comprising:

(a) at least one enzyme of the laccase type;

(b) at least one alkalinizing agent chosen from:

(i) basic amino acids;

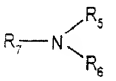
(ii) compounds of the following formula (A):

$$X(OH)_n$$

wherein n is equal to 1 or 2; X is chosen from K, Li and $N^+R_1R_2R_3R_4$ wherein R_1, R_2, R_3, and R_4, which are identical or different, are each chosen from C_1-C_4 alkyl groups, C_1-C_4 monohydroxyalkyl groups and C_2-C_4 polyhydroxyalkyl groups when n=1; or

X is chosen from Mg and Ca when n=2;

(iii) compounds of the following formula (B):



wherein R_5 is chosen from hydrogen groups, C_1-C_6 alkyl groups, C_1-C_6 monohydroxyalkyl groups, and C_2-C_6 polyhydroxyalkyl groups;

R_6 and R_7, which are identical or different, are each chosen from hydrogen groups, C_1-C_6 alkyl groups, C_1-C_6 monohydroxyalkyl groups, and C_2-C_6 polyhydroxyalkyl groups;

with the proviso that R_5, R_6, and R_7 are not simultaneously chosen from C_2 β-hydroxyalkyl groups;

with the additional proviso that if R_6 and R_7 are simultaneously chosen from H, then R_5 is not chosen from C_2 monohydroxyalkyl groups and branched C_4 monohydroxyalkyl groups;

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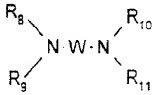
and with the additional proviso that if R_5 is chosen from H and C_1-C_6 alkyl groups and simultaneously R_6 is chosen from C_1-C_6 alkyl groups, then R_7 is not chosen from H and C_1-C_6 alkyl groups; and

(iv) compounds of the following formula (C):



(C)

wherein W is chosen from propylene groups optionally substituted with a substituent chosen from hydroxyl groups and C_1-C_4 alkyl groups; R_8, R_9, R_{10} and R_{11}, which are identical or different, are each chosen from hydrogen groups, C_1-C_4 alkyl groups and C_1-C_4 hydroxyalkyl groups; and

(c) at least one oxidation dye with the proviso that said at least one oxidation dye is not chosen from autooxidizable indole dyes.

59. A method of dyeing keratinous fibers according to Claim 58, wherein said keratinous fibers are human keratinous fibers.

60. A method of dyeing keratinous fibers according to Claim 59, wherein said human keratinous fibers are hair.

61. A method for dyeing keratinous fibers comprising the steps of:

(a) storing a first composition,

(b) storing a second composition separately from said first composition,

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(c) mixing the first composition with the second composition to form a mixture, and (d) applying said mixture to said keratinous fibers for a time sufficient to achieve a desired colouration,

wherein said first composition comprises said at least one oxidation dye in a medium appropriate for keratinous fibers, and

wherein said second composition comprises said at least one enzyme of the laccase type and said at least one alkalinizing agent in a medium appropriate for keratinous fibers.

62. A multicompartment device or a dyeing kit, comprising a first compartment containing a composition (A) comprising, in a medium appropriate for dyeing, at least one oxidation dye and a second compartment containing a composition (B), comprising, in a medium appropriate for keratinous fibers, at least one enzyme of the laccase type, wherein at least one of said composition (A) and composition (B) comprises at least one alkalinizing agent chosen from:

(i) basic amino acids;

(ii) compounds of the following formula (A):

$$X(OH)_n$$

wherein n is equal to 1 or 2; X is chosen from K, Li and $N^+R_1R_2R_3R_4$

wherein R_1, R_2, R_3, and R_4, which are identical or different, are each

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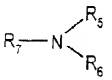
chosen from C_1-C_4 alkyl groups, C_1-C_4 monohydroxyalkyl groups and C_2-C_4 polyhydroxyalkyl groups when n=1; or

X is chosen from Mg and Ca when n=2;

(iii) compounds of the following formula (B):



wherein R_5 is chosen from hydrogen groups, C_1-C_6 alkyl groups, C_1-C_6 monohydroxyalkyl groups, and C_2-C_6 polyhydroxyalkyl groups;

R_6 and R_7, which are identical or different, are each chosen from hydrogen groups, C_1-C_6 alkyl groups, C_1-C_6 monohydroxyalkyl groups, and C_2-C_6 polyhydroxyalkyl groups;

with the proviso that R_5, R_6, and R_7 are not simultaneously chosen from C_2 β-hydroxyalkyl groups;

with the additional proviso that if R_6 and R_7 are simultaneously chosen from H, then R_5 is not chosen from C_2 monohydroxyalkyl groups and branched C_4 monohydroxyalkyl groups;

and with the additional proviso that if R_5 is chosen from H and C_1-C_6 alkyl groups

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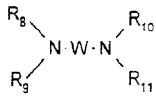
and simultaneously R_6 is chosen from C_1-C_6 alkyl groups, then R_7 is not chosen from H and C_1-C_6 alkyl groups; and

(iv) compounds of the following formula (C):



(C)

wherein W is chosen from propylene groups optionally substituted with a substituent chosen from hydroxyl groups and C_1-C_4 alkyl groups; R_8, R_9, R_{10} and R_{11}, which are identical or different, are each chosen from hydrogen groups, C_1-C_4 alkyl groups and C_1-C_4 hydroxyalkyl groups.--

REMARKS

Claims 1-22 are canceled. Claims 23-62 have been added, and are fully supported by the original application disclosure. No new matter has been added.

If the Examiner believes a telephone conference would be helpful in advancing the prosecution of this application, the Examiner is respectfully urged to contact Applicants' undersigned representative at (202) 408-4082.

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Please grant any extensions of time required to enter this response and charge any additional required fees to our deposit account 06-0916.

Respectfully submitted,

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DYEING COMPOSITION CONTAINING A LACCASE AND METHOD FOR DYEING KERATINOUS FIBRES

The present invention relates to a composition for the oxidation dyeing of keratinous fibres comprising at least one enzyme of the laccase type, at least one oxidation dye and at least one particular alkalinizing agent as well as the methods of dyeing keratinous fibres, in particular human hair using this composition.

It is known to dye keratinous fibres, and in particular human hair, with dyeing compositions containing oxidation dye precursors, in particular ortho- and para-phenylenediamines, ortho- or para-aminophenols, heterocyclic bases generally called oxidation bases. The oxidation dye precursors, or oxidation bases, are colourless or weakly coloured compounds which, combined with oxidizing products, can give rise to coloured compounds by a process of oxidative condensation.

It is also known that the shades obtained with these oxidation bases can be varied by combining them with couplers or colour modifiers, the latter being chosen in particular from aromatic meta-diamines, meta-aminophenols, meta-diphenols and certain heterocyclic compounds.

The variety of molecules used in oxidation bases and couplers allows a rich palette of colours to be obtained.

The so-called "permanent" colour obtained by means of these oxidation dyes should moreover satisfy a number of requirements. Thus, it should have no drawbacks from the toxicological point of view, it should make it possible to obtain shades of the desired intensity and it should exhibit good resistance towards external agents (light, adverse weather conditions, washing, permanent waving, perspiration, rubbing).

The dyes should also make it possible to cover grey hair, and thus should be the least selective possible, that is to say they should make it possible to obtain the smallest possible differences in colour all along the same keratinous fibre, which may indeed be differently sensitized (i.e. damaged) between its tip and its root.

The oxidation dyeing of keratinous fibres is generally carried out in an alkaline medium, in the presence of hydrogen peroxide. However, the use of alkaline media in the presence of hydrogen peroxide has the disadvantage of causing substantial degradation of the fibres, as well as decolouring of the keratinous fibres which is not always desirable.

The oxidation dyeing of keratinous fibres can also be carried out with the aid of oxidizing systems different from hydrogen peroxide such as enzymatic systems. Thus, it has already been proposed in Patent US 3,251,742, Patent Applications FR-A-2,112,549, FR-A-2,694,018, EP-A-0,504,005, WO95/07988, WO95/33836,

WO95/33837, WO96/00290, WO97/19998 and WO97/19999 to dye keratinous fibres with compositions comprising at least one oxidation dye in combination with enzymes of the laccase type, the said compositions being brought into contact with atmospheric oxygen. These dyeing formulations, although used under conditions which do not cause degradation of the keratinous fibres comparable to that caused by dyeings carried out in the presence of hydrogen peroxide, lead to colours which are still inadequate both from the point of view of homogeneity of the colour distributed along the fibre ("unison"), from the point of view of chromaticity (luminosity) and of the dyeing power.

The aim of the present invention is to solve the problems mentioned above.

The applicant has surprisingly discovered novel compositions containing, as oxidizing system, at least one enzyme of the laccase type and at least one particular alkalinizing agent which will be defined in greater detail below, which may constitute, in the presence of oxidation dyes, ready-to-use dyeing formulations leading to colours which are more homogeneous, more intense and more chromatic without causing significant degradation or decolouring of the keratinous fibres, exhibiting low selectivity and good resistance to various attacks to which the hair may be subjected.

These discoveries form the basis of the present invention.

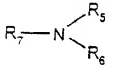
The first subject of the present invention is therefore a ready-to-use composition intended for the oxidation dyeing of keratinous fibres, in particular human keratinous fibres and more particularly human hair, comprising, in a carrier appropriate for dyeing keratinous fibres:

- (a) at least one enzyme of the laccase type;
- (b) at least one alkalinizing agent chosen from the group consisting of:

(i) a basic amino acid;

(ii) a compound of the following formula (A): $X(OH)_n$ in which X represents K, Li when n=1; X represents Mg, Ca when n=2; X represents $N^+R_1R_2R_3R_4$ with R_1, R_2, R_3, R_4, which are identical or different, denoting a C_1-C_4 alkyl radical, a C_1-C_4 monohydroxyalkyl or C_2-C_4 polyhydroxyalkyl radical, when n=1;

(iii) a compound of the following formula (B):



in which R_5 denotes a C_1-C_6 alkyl radical, a C_1-C_6 monohydroxyalkyl or C_2-C_6 polyhydroxyalkyl radical; R_6, R_7, which are identical or different, denote a hydrogen atom, a C_1-C_6 alkyl radical, a C_1-C_6 monohydroxyalkyl or C_2-C_6 polyhydroxyalkyl radical;

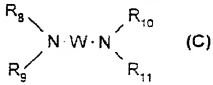
with the proviso that

• R_5, R_6, R_7 do not simultaneously denote the C_2 β-hydroxyalkyl radical,

• if R_6 and R_7 simultaneously denote H, then R_5 does not denote a C_2 monohydroxyalkyl or branched C_4 monohydroxyalkyl radical,

• if R_5 denotes hydrogen or a C_1-C_6 alkyl radical and at the same time R_6 denotes a C_1-C_6 alkyl radical, then R_7 does not denote H or a C_1-C_6 alkyl radical;

(iv) a compound of the following formula (C):



(C)

in which W is a propylene residue optionally substituted with a hydroxyl group or a C_1-C_4 alkyl radical; R_8, R_9, R_{10} and R_{11}, which are identical or different, represent a hydrogen atom, a C_1-C_4 alkyl or C_1-C_4 hydroxyalkyl radical;

-(c) at least one oxidation dye with the exception of autooxidizable indole dyes.

The laccase(s) used in the ready-to-use dye composition in accordance with the invention may be chosen in particular from laccases of plant origin, animal origin, fungal origin (yeasts, moulds, fungi) or bacterial origin, organisms which may be of mono- or pluricellular origin. They can be obtained by biotechnology.

Among the laccases of plant origin which can be used according to the invention, there may be

mentioned the laccases produced by plants which perform chlorophyll synthesis as indicated in Application FR-A-2,694,018 such as those found in the extracts of Anacardiaceae such as for example the extracts of Magnifera indica, Schinus molle or Pleiogynium timoriense, in the extracts of Podocarpaceae, Rosmarinus off., Solanum tuberosum, Iris sp., Coffea sp., Daucus carrota, Vinca minor, Persea americana, Catharenthus roseus, Musa sp., Malus pumila, Gingko biloba, Monotropa hypopithys (Indian pipe), Aesculus sp., Acer pseudoplatanus, Prunus persica, Pistacia palaestina.

Among the laccases of fungal origin optionally obtained by biotechnology which can be used according to the invention, there may be mentioned the laccase(s) derived from Polyporus versicolor, Rhizoctonia practicola and Rhus vernicifera as indicated in Applications FR-A-2,112,549 and EP-A-504005, those described in Patent Application WO95/07988, WO95/33836, WO95/33837, WO96/00290, WO97/19998 and WO97/19999, whose content is an integral part of the present description, such as for example those derived from Scytalidium, Polyporus pinsitus, Myceliophtora thermophila, Rhizoctonia solani, Pyricularia orizae, or variants thereof. There may also be mentioned those derived from Tramates versicolor, Fomes fomentarius, Chaetomium thermophile, Neurospora crassa, Coriolus versicol, Botrytis cinerea,

Rigidoporus lignosus, Phellinus noxius, Pleurotus ostreatus, Aspergillus nidulans, Podospora anserina, Agaricus bisporus, Ganoderma lucidum, Glomerella cingulata, Lactarius piperatus, Russula delica, Heterobasidion annosum, Thelephora terrestris, Cladosporium cladosporiodes, Cerrena unicolor, Coriolus hirsutus, Ceriporiopsis subvermispora, Coprinus cinereus, Panaeolus papilionaceus, Panaeolus sphinctrinus, Schizophyllum commune, Dichomitius squalens and variants thereof.

The laccases of fungal origin optionally obtained by biotechnology will be preferably chosen.

The enzymatic activity of the laccases of the invention which have syringaldazine among their substrates can be defined from the oxidation of syringaldazine under aerobic conditions. The lacu unit corresponds to the quantity of enzyme catalysing the conversion of 1 mmol of syringaldazine per minute at pH 5.5 at 30°C. The unit u corresponds to the quantity of enzyme producing a delta absorbance at 530 nm of 0.001 per minute using syringaldazine as substrate, at 30°C and at pH 6.5.

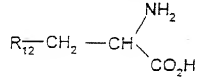
The enzymatic activity of the laccases of the invention can also be defined from the oxidation of para-phenylenediamine. The lacu unit corresponds to the quantity of enzyme producing a delta absorbance at 496.5 nm of 0.001 per minute using para-phenylenediamine as substrate (64 mM) at 30°C and at

pH 5. According to the invention, it is preferable to determine the enzymatic activity in lacu units.

The quantities of laccase used in the compositions of the invention will vary according to the nature of the laccase chosen. Preferably, they will vary from 0.5 to 2000 lacu, or from 1000 to 4×10^7 u units, or from 20 to 2×10^6 lacu units per 100 g of composition.

For the purposes of the present invention, i.e. in the preceding text and in the text which follows, "basic amino acid" defines either (i) an amino acid having, in addition to the amine function positioned in α with respect to the carboxyl group, an additional cationic (or basic) group; or (ii) an amino acid having a cationic (or basic) (hydrophilic) side chain; or (iii) an amino acid carrying a side chain consisting of a nitrogenous base. These definitions are generally known and are published in general biochemistry books such as J.H. WEIL (1983) pages 5 and the following pages, Lubert STRYER (1995) page 22, A. LEHNINGER (1993) pages 115-116, DE BOECK-WESMAEL (1994) pages 57-59.

The basic amino acids in accordance with the invention are preferably chosen from those corresponding to the following formula (D):



(D)

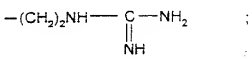
where R_{12} denotes a group chosen from:

;

$-(CH_2)_3NH_2$;

$-(CH_2)_2NH_2$

$-(CH_2)_2NHCONH_2$;



;

The compounds corresponding to formula (D) are histidine, lysine, ornithine, citrulline, arginine.

In the compounds of formula (A) or (B), according to the invention, the alkyl radicals may be linear or branched and the polyhydroxyalkyl radicals denote radicals comprising from 2 to 6 hydroxyl groups and preferably from 2 to 4.

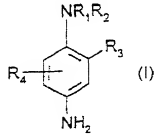
The compounds of formula (B), according to the invention, are preferably chosen from the group consisting of diethanolamine, monoisopropanolamine, diisopropanolamine, triisopropanolamine, 2-amino-2-methyl-1,3-propanediol, 2-amino-2-ethyl-1,3-propanediol, 2-amino-1-n-butanol, 1-diethylamino-2,3-propanediol, tris(hydroxymethyl)aminomethane, ethylmonoethanolamine.

The compositions in accordance with the invention contain the particular alkalinizing agents defined above in contents by weight which may range from 0.001% to 20%, preferably from 0.01% to 5% and still more preferably from 0.05% to 3%, relative to the total weight of the composition.

The nature of the oxidation dye(s) used in the ready-to-use dyeing composition is not critical. They are chosen from oxidation bases and/or couplers.

The oxidation bases may be chosen in particular from para-phenylenediamines, double bases, para-aminophenols, ortho-aminophenols and heterocyclic oxidation bases.

Among the para-phenylenediamines which can be used as oxidation base in the dyeing composition in accordance with the invention, there may be mentioned in particular the compounds of the following formula (I) and their addition salts with an acid:



(I)

in which:

- R_1 represents a hydrogen atom, a C_1-C_4 alkyl radical, a monohydroxy(C_1-C_4 alkyl) radical, a polyhydroxy-(C_2-C_4 alkyl) radical, a (C_1-C_4)alkoxy(C_1-C_4)alkyl radical, a C_1-C_4 alkyl radical substituted with a

nitrogen-containing group, a phenyl radical or a 4'-aminophenyl radical;

- R_2 represents a hydrogen atom, a C_1-C_4 alkyl radical, a monohydroxy(C_1-C_4 alkyl) radical, a polyhydroxy(C_2-C_4 alkyl) radical, a (C_1-C_4)alkoxy(C_1-C_4)alkyl radical or a C_1-C_4 alkyl radical substituted with a nitrogen-containing group;
- R_3 represents a hydrogen atom, a halogen atom such as a chlorine, bromine, iodine or fluorine atom, a C_1-C_4 alkyl radical, a monohydroxy(C_1-C_4 alkyl) radical, a hydroxy(C_1-C_4 alkoxy) radical, an acetylamino(C_1-C_4 alkoxy) radical, a mesylamino(C_1-C_4 alkoxy) radical or a carbamoylamino(C_1-C_4 alkoxy) radical,
- R_4 represents a hydrogen or halogen atom or a C_1-C_4 alkyl radical.

Among the nitrogen-containing groups of formula (I) above, there may be mentioned in particular the amino, mono(C_1-C_4)alkylamino, (C_1-C_4)dialkylamino, (C_1-C_4)trialkylamino, monohydroxy(C_1-C_4)alkylamino, imidazolinium and ammonium radicals.

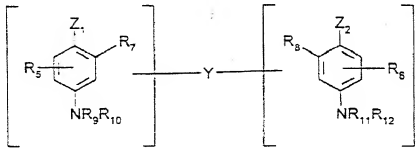
Among the para-phenylenediamines of formula (I) above, there may be mentioned more particularly para-phenylenediamine, para-tolylenediamine, 2-chloro-para-phenylenediamine, 2,3-dimethyl-para-phenylene-diamine, 2,6-dimethyl-para-phenylenediamine, 2,6-diethyl-para-phenylenediamine, 2,5-dimethyl-para-phenylenediamine, N,N-dimethyl-para-phenylenediamine, N,N-diethyl-para-phenylenediamine, N,N-dipropyl-para-

phenylenediamine, 4-amino-N,N-diethyl-3-methylaniline, N,N-bis(β-hydroxyethyl)-para-phenylenediamine, 4-N,N-bis(β-hydroxyethyl)amino-2-methylaniline, 4-N,N-bis(β-hydroxyethyl)amino-2-chloroaniline, 2-β-hydroxyethyl-para-phenylenediamine, 2-fluoro-para-phenylenediamine, 2-isopropyl-para-phenylenediamine, N-(β-hydroxypropyl)-para-phenylenediamine, 2-hydroxymethyl-para-phenylenediamine, N,N-dimethyl-3-methyl-para-phenylenediamine, N,N-(ethyl-β-hydroxyethyl)-para-phenylenediamine, N-(β,γ-dihydroxypropyl)-para-phenylenediamine, N-(4'-aminophenyl)-para-phenylenediamine, N-phenyl-para-phenylenediamine, 2-β-hydroxyethyloxy-para-phenylenediamine, 2-β-acetylaminoethyloxy-para-phenylenediamine, N-(β-methoxyethyl)-para-phenylenediamine, and their addition salts with an acid.

Among the para-phenylenediamines of formula (I) above, there are most particularly preferred para-phenylenediamine, para-tolylenediamine, 2-isopropyl-para-phenylenediamine, 2-β-hydroxyethyl-para-phenylenediamine, 2-β-hydroxyethyloxy-para-phenylenediamine, 2,6-dimethyl-para-phenylenediamine, 2,6-diethyl-para-phenylenediamine, 2,3-dimethyl-para-phenylenediamine, N,N-bis(β-hydroxyethyl)-para-phenylenediamine, 2-chloro-para-phenylenediamine, 2-β-acetylaminoethyloxy-para-phenylenediamine, and their addition salts with an acid.

According to the invention, "double bases" is understood to mean the compounds containing at least two aromatic rings on which amino and/or hydroxyl groups are carried.

Among the double bases which can be used as oxidation bases in the dyeing compositions in accordance with the invention, there may be mentioned in particular the compounds corresponding to the following formula (II), and their addition salts with an acid:



(II)

in which:

- Z_1 and Z_2, which are identical or different, represent a hydroxyl or $-NH_2$ radical which may be substituted with a C_1-C_4 alkyl radical or with a linking arm Y;

- the linking arm Y represents a linear or branched alkylene chain comprising from 1 to 14 carbon atoms, which may be interrupted by or which may end with one or more nitrogen-containing groups and/or one or more heteroatoms such as oxygen, sulphur or nitrogen atoms, and optionally substituted with one or more hydroxyl or C_1-C_6 alkoxy radicals;

- R_5 and R_6 represent a hydrogen or halogen atom, a C_1-C_4 alkyl radical, a monohydroxy(C_1-C_4 alkyl) radical, a polyhydroxy(C_2-C_4 alkyl) radical, an amino(C_1-C_4 alkyl) radical or a linking arm Y;
- R_7, R_8, R_9, R_{10}, R_{11} and R_{12}, which are identical or different, represent a hydrogen atom, a linking arm Y or a C_1-C_4 alkyl radical;

it being understood that the compounds of formula (II) contain only one linking arm Y per molecule.

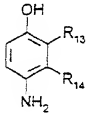
Among the nitrogen-containing groups of formula (II) above, there may be mentioned in particular the amino, mono(C_1-C_4)alkylamino, (C_1-C_4)dialkylamino, (C_1-C_4)trialkylamino, monohydroxy(C_1-C_4)alkylamino, imidazolinium and ammonium radicals.

Among the double bases of formulae (II) above, there may be mentioned more particularly N,N'-bis(β-hydroxyethyl)-N,N'-bis(4'-aminophenyl)-1,3-diaminopropanol, N,N'-bis(β-hydroxyethyl)-N,N'-bis(4'-aminophenyl)ethylenediamine, N,N'-bis(4-aminophenyl)-tetramethylenediamine, N,N'-bis(β-hydroxyethyl)-N,N'-bis(4-aminophenyl)tetramethylenediamine, N,N'-bis(4-methylaminophenyl)tetramethylenediamine, N,N'-bis(ethyl)-N,N'-bis(4'-amino-3'-methylphenyl)ethylene-diamine, 1,8-bis(2,5-diaminophenoxy)-3,5-dioxaoctane, and their addition salts with an acid.

Among these double bases of formula (II), N,N'-bis(β-hydroxyethyl)-N,N'-bis(4'-aminophenyl)-1,3-

diaminopropanol, 1,8-bis(2,5-diaminophenoxy)-3,5-dioxaoctane or one of their addition salts with an acid are particularly preferred.

Among the para-aminophenols which can be used as oxidation bases in the dyeing compositions in accordance with the invention, there may be mentioned in particular the compounds corresponding to the following formula (III), and their addition salts with an acid:



(III)

in which:

- R_{13} represents a hydrogen or halogen atom, a C_1-C_4 alkyl, monohydroxy(C_1-C_4 alkyl), (C_1-C_4)alkoxy(C_1-C_4)-alkyl, amino(C_1-C_4 alkyl) or hydroxy(C_1-C_4)alkylamino-(C_1-C_4 alkyl) radical,
- R_{14} represents a hydrogen or halogen atom, a C_1-C_4 alkyl, monohydroxy(C_1-C_4 alkyl), polyhydroxy(C_2-C_4 alkyl), amino(C_1-C_4 alkyl), cyano(C_1-C_4 alkyl) or (C_1-C_4)alkoxy(C_1-C_4)alkyl radical,

it being understood that at least one of the radicals R_{13} or R_{14} represents a hydrogen atom.

Among the para-aminophenols of formula (III) above, there may be mentioned more particularly para-aminophenol, 4-amino-3-methylphenol, 4-amino-3-fluorophenol, 4-amino-3-hydroxymethylphenol, 4-amino-2-

methylphenol, 4-amino-2-hydroxymethylphenol, 4-amino-2-methoxymethylphenol, 4-amino-2-aminomethylphenol, 4-amino-2-(β-hydroxyethylaminomethyl)phenol, 4-amino-2-fluorophenol, and their addition salts with an acid.

Among the ortho-aminophenols which can be used as oxidation bases in the dyeing compositions in accordance with the invention, there may be mentioned more particularly 2-aminophenol, 2-amino-5-methylphenol, 2-amino-6-methylphenol, 5-acetamido-2-aminophenol, and their addition salts with an acid.

Among the heterocyclic bases which can be used as oxidation bases in the dyeing compositions in accordance with the invention, there may be mentioned more particularly pyridine derivatives, pyrimidine derivatives, pyrazole derivatives, pyrazolopyrimidine derivatives, and their addition salts with an acid.

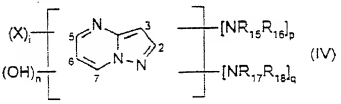
Among the pyridine derivatives, there may be mentioned more particularly the compounds described for example in Patents GB 1,026,978 and GB 1,153,196, such as 2,5-diaminopyridine, 2-(4-methoxyphenyl)amino-3-aminopyridine, 2,3-diamino-6-methoxypyridine, 2-(β-methoxyethyl)amino-3-amino-6-methoxypyridine, 3,4-diaminopyridine, and their addition salts with an acid.

Among the pyrimidine derivatives, there may be mentioned more particularly the compounds described for example in German Patent DE 2,359,399 or Japanese Patents JP 88-169,571 and JP 91-333,495 or Patent

Application WO 96/15765, such as 2,4,5,6-tetra-aminopyrimidine, 4-hydroxy-2,5,6-triaminopyrimidine, 2-hydroxy-4,5,6-triaminopyrimidine, 2,4-dihydroxy-5,6-diaminopyrimidine, 2,5,6-triaminopyrimidine, and their addition salts with an acid.

Among the pyrazole derivatives, there may be mentioned more particularly the compounds described in Patents DE 3,843,892, DE 4,133,957 and Patent Applications WO 94/08969, WO 94/08970, FR-A-2,733,749 and DE 195 43 988 such as 4,5-diamino-1-methylpyrazole, 3,4-diaminopyrazole, 4,5-diamino-1-(4'-chlorobenzyl)-pyrazole, 4,5-diamino-1,3-dimethylpyrazole, 4,5-diamino-3-methyl-1-phenylpyrazole, 4,5-diamino-1-methyl-3-phenylpyrazole, 4-amino-1,3-dimethyl-5-hydrazinopyrazole, 1-benzyl-4,5-diamino-3-methyl-pyrazole, 4,5-diamino-3-tert-butyl-1-methylpyrazole, 4,5-diamino-1-tert-butyl-3-methylpyrazole, 4,5-diamino-1-(β-hydroxyethyl)-3-methylpyrazole, 4,5-diamino-1-ethyl-3-methylpyrazole, 4,5-diamino-1-ethyl-3-(4'-methoxyphenyl)pyrazole, 4,5-diamino-1-ethyl-3-hydroxymethylpyrazole, 4,5-diamino-3-hydroxymethyl-1-methylpyrazole, 4,5-diamino-3-hydroxymethyl-1-isopropylpyrazole, 4,5-diamino-3-methyl-1-isopropyl-pyrazole, 4-amino-5-(2'-aminoethyl)amino-1,3-dimethyl-pyrazole, 3,4,5-triaminopyrazole, 1-methyl-3,4,5-triaminopyrazole, 3,5-diamino-1-methyl-4-methylamino-pyrazole, 3,5-diamino-4-(β-hydroxyethyl)amino-1-methylpyrazole, and their addition salts with an acid.

Among the pyrazolopyrimidine derivatives, there may be mentioned more particularly the pyrazolo[1,5-a]pyrimidines of the following formula (IV), their addition salts with an acid or with a base and their tautomeric forms, when a tautomeric equilibrium exists:



(IV)

in which:

- R_{15}, R_{16}, R_{17} and R_{18}, which are identical or different, denote a hydrogen atom, a C_1-C_4 alkyl radical, an aryl radical, a C_1-C_4 hydroxyalkyl radical, a C_2-C_4 polyhydroxyalkyl radical, a (C_1-C_4)alkoxy(C_1-C_4 alkyl) radical, a C_1-C_4 aminoalkyl radical (it being possible for the amine to be protected with an acetyl, ureido or sulphonyl radical), a (C_1-C_4)alkylamino(C_1-C_4 alkyl) radical, a di-[(C_1-C_4)alkyl]amino(C_1-C_4 alkyl) radical (it being possible for the dialkyl radicals to form a carbon-containing ring or a 5- or 6-membered heterocycle), a hydroxy(C_1-C_4)alkyl- or di-[hydroxy(C_1-C_4)alkyl]-amino(C_1-C_4 alkyl) radical,
- the X radicals, which are identical or different, denote a hydrogen atom, a C_1-C_4 alkyl radical, an aryl radical, a C_1-C_4 hydroxyalkyl radical, a C_2-C_4 polyhydroxyalkyl radical, a C_1-C_4 aminoalkyl radical,

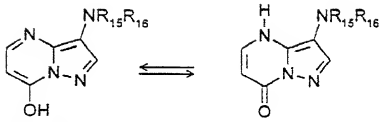
a (C_1-C_4)alkylamino(C_1-C_4 alkyl) radical, a di-[(C_1-C_4)alkyl]amino(C_1-C_4 alkyl) radical (it being possible for the dialkyls to form a carbon-containing ring or a 5- or 6-membered heterocycle), a hydroxy(C_1-C_4)alkyl or di-[hydroxy(C_1-C_4)alkyl]-amino(C_1-C_4 alkyl) radical, an amino radical, a (C_1-C_4)alkyl- or di-[(C_1-C_4)alkyl]-amino radical; a halogen atom, a carboxylic acid group, a sulphonic acid group;

- i equals 0, 1, 2 or 3;
- p equals 0 or 1;
- q equals 0 or 1;
- n equals 0 or 1;

with the proviso that:

- the sum p + q is different from 0;
- when p + q is equal to 2, then n equals 0 and the groups $NR_{15}R_{16}$ and $NR_{17}R_{18}$ occupy positions (2,3); (5,6); (6,7); (3,5) or (3,7);
- when p + q is equal to 1, then n equals 1 and the group $NR_{15}R_{16}$ (or $NR_{17}R_{18}$) and the OH group occupy positions (2,3); (5,6); (6,7); (3,5) or (3,7).

When the pyrazolo[1,5-a]pyrimidines of formula (IV) above are such that they comprise a hydroxyl group on one of the positions 2, 5 or 7 at the α position with respect to a nitrogen atom, a tautomeric equilibrium exists which is represented for example by the following scheme:



Among the pyrazolo[1,5-a]pyrimidines of formula (IV) above, there may be mentioned in particular:

- pyrazolo[1,5-a]pyrimidine-3,7-diamine;
- 2,5-dimethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;
- pyrazolo[1,5-a]pyrimidine-3,5-diamine;
- 2,7-dimethylpyrazolo[1,5-a]pyrimidine-3,5-diamine;
- 3-aminopyrazolo[1,5-a]pyrimidin-7-ol;
- 3-aminopyrazolo[1,5-a]pyrimidin-5-ol;
- 2-(3-aminopyrazolo[1,5-a]pyrimidin-7-ylamino)ethanol;
- 2-(7-aminopyrazolo[1,5-a]pyrimidin-3-ylamino)ethanol;
- 2-[(3-aminopyrazolo[1,5-a]pyrimidin-7-yl)(2-hydroxyethyl)amino]ethanol;
- 2-[(7-aminopyrazolo[1,5-a]pyrimidin-3-yl)(2-hydroxyethyl)amino]ethanol;
- 5,6-dimethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;
- 2,6-dimethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;
- 2,5,N7,N7-tetramethylpyrazolo[1,5-a]pyrimidine-3,7-diamine;

and their addition salts and their tautomeric forms, when a tautomeric equilibrium exists.

The pyrazolo[1,5-a]pyrimidines of formula (IV) above may be prepared by cyclization from an

aminopyrazole according to the syntheses described in the following references:

- EP 628559 BEIERSDORF-LILLY
- R. Vishdu, H. Navedul, Indian J. Chem., 34b(6), 514, 1995.
- N.S. Ibrahim, K.U. Sadek, F.A. Abdel-Al, Arch. Pharm., 320, 240, 1987.
- R.H. Springer, M.B. Scholten, D.E. O'Brien, T. Novinson, J.P. Miller, R.K. Robins, J. Med. Chem., 25, 235, 1982.
- T. Novinson, R.K. Robins, T.R. Matthews, J. Med. Chem., 20, 296, 1977.
- US 3907799 ICN PHARMACEUTICALS

The pyrazolo[1,5-a]pyrimidines of formula (IV) above can also be prepared by cyclization from hydrazine according to the syntheses described in the following references:

- A. McKillop and R.J. Kobilecki, Heterocycles, 6(9), 1355, 1977.
- E. Alcade, J. De Mendoza, J.M. Marcia-Marquina, C. Almera, J. Elguero, J. Heterocyclic Chem., 11(3), 423, 1974.
- K. Saito, I. Hori, M. Higarashi, H. Midorikawa, Bull. Chem. Soc. Japan, 47(2), 476, 1974.

The oxidation base(s) preferably represent from 0.0005 to 12% by weight approximately of the total weight of the dyeing composition in accordance with the

invention, and still more preferably from 0.005 to 6% by weight approximately of this weight.

The coupler(s) which can be used in the ready-to-use dyeing composition in accordance with the invention are those conventionally used in oxidation dyeing compositions, that is to say meta-phenylene-diamines, meta-aminophenols, meta-diphenols, heterocyclic couplers, and their addition salts with an acid.

These couplers may be chosen in particular from 2-methyl-5-aminophenol, 5-N-(β-hydroxyethyl)amino-2-methylphenol, 3-aminophenol, 1,3-dihydroxybenzene, 1,3-dihydroxy-2-methylbenzene, 4-chloro-1,3-dihydroxy-benzene, 2,4-diamino-1-(β-hydroxyethyloxy)benzene, 2-amino-4-(β-hydroxyethylamino)-1-methoxybenzene, 1,3-diaminobenzene, 1,3-bis(2,4-diaminophenoxy)propane, sesamol, α-naphthol, 6-hydroxyindole, 4-hydroxyindole, 4-hydroxy-N-methylindole, 6-hydroxyindoline, 2,6-dihydroxy-4-methylpyridine, 1-H-3-methylpyrazol-5-one, 1-phenyl-3-methylpyrazol-5-one, 2,6-dimethyl-pyrazolo[1,5-b]-1,2,4-triazole, 2,6-dimethyl[3,2-c]-1,2,4-triazole, 6-methylpyrazolo[1,5-a]benzimidazole, and their addition salts with an acid.

These couplers preferably represent from 0.0001 to 10% by weight approximately of the total weight of the ready-to-use dyeing composition, and still more preferably from 0.005 to 5% by weight approximately of this weight.

In general, the addition salts with an acid which can be used in the context of the dyeing compositions of the invention (oxidation bases and couplers) are in particular chosen from hydrochlorides, hydrobromides, sulphates and tartrates, lactates and acetates.

The dyeing composition of the invention may also contain, in addition to the oxidation dyes defined above, direct dyes in order to increase the shimmer of the shades. These direct dyes can then in particular be chosen from nitro, azo or anthraquinone dyes.

The ready-to-use dyeing composition in accordance with the invention may also contain various adjuvants conventionally used in hair dyeing compositions, such as anionic, cationic, nonionic, amphoteric or zwitterionic surfactants or mixtures thereof, polymers, thickeners, antioxidants, enzymes different from the laccases used in accordance with the invention, such as for example peroxidases or oxido-reductases containing 2 electrons, penetrating agents, sequestering agents, perfumes, dispersing agents, film-forming agents, screening agents, vitamins, preservatives or opacifying agents.

Of course, persons skilled in the art will be careful to choose this or these optional additional compounds such that the advantageous properties intrinsically attached to the ready-to-use dyeing composition in accordance with the invention are not,

or substantially not, impaired by the addition(s) envisaged.

The ready-to-use dyeing composition in accordance with the invention can be provided in various forms, such as in the form of liquids, creams, gels, optionally pressurized, or in any other form appropriate for dyeing keratinous fibres, in particular human hair. In this case, the oxidation dye(s) and the laccase(s) are present in the same ready-to-use composition, and consequently the said composition should be free of gaseous oxygen, so as to avoid any premature oxidation of the oxidation dye(s).

The subject of the invention is also a method of dyeing keratinous fibres, and in particular human keratinous fibres such as hair, using the ready-to-use dyeing composition as defined above.

According to this method, at least one ready-to-use dyeing composition as defined above is applied to the fibres for a sufficient time to develop the desired colour, after which they are rinsed, optionally washed with shampoo, rinsed again and dried.

The time necessary for the development of the colour on the keratinous fibres is generally between 3 and 60 minutes and still more precisely 5 and 40 minutes.

According to one particular embodiment of the invention, the method comprises a preliminary step consisting in storing in a separate form, on the one

hand, a composition (A) comprising, in a medium appropriate for dyeing, at least one oxidation dye as defined above and, on the other hand, a composition (B) containing, in a medium appropriate for dyeing, at least one enzyme of the laccase type and at least one particular alkalinizing agent as defined above, and then in mixing them at the time of use before applying this mixture to the keratinous fibres.

According to a specific embodiment of the invention, the alkalinizing agent may be incorporated into the composition (A).

Another subject of the invention is a multi-compartment device or dyeing (kit) or any other multi-compartment packaging system in which a first comparment contains the composition (A) as defined above and a second compartment contains a composition (B) as defined above. These devices may be equipped with a means which makes it possible to deliver the desired mixture to the hair, such as the devices described in Patent FR-2,586,913 in the name of the applicant.

The medium appropriate for keratinous fibres (or carrier) of the dyeing compositions in accordance with the invention generally consists of water or of a mixture of water and of at least one organic solvent in order to solubilize the compounds which might not be sufficiently soluble in water. As organic solvent, there may be mentioned for example C_1-C_4 alkanols such

as ethanol and isopropanol as well as aromatic alcohols such as benzyl alcohol, similar products and mixtures thereof.

The solvents may be present in proportions preferably of between 1 and 40% by weight approximately relative to the total weight of the dyeing composition, and still more preferably between 5 and 30% by weight approximately.

The pH of the dyeing compositions in accordance with the invention is chosen such that the enzymatic activity of the laccase is not impaired. It varies generally from 6 to 11 approximately, and more preferably from 6 to 9 approximately.

Concrete examples illustrating the invention will now be given.

In the text which follows and in the preceding text, unless otherwise stated, the percentages are expressed by weight.

The examples below illustrate the invention with no limitation being implied.

EXAMPLE 1 **Dyeing composition**

The following ready-to-use dyeing composition was prepared (contents in grams):

- Laccase obtained from Rhus vernicifera containing 180 units/mg units/mg, marketed by the company SIGMA 1.8 g
- (C_8-C_{10})Alkyl polyglucoside in aqueous solution containing 60% of active 8.0 g

substance (AS) sold under the name ORAMIX CG110 by the company SEPPIC

- Paraphenylenediamine 0.254 g
- 2,4-Diaminophenoxyethanol dihydrochloride 0.260 g
- Arginine qs pH 6.5
- Demineralized water qs 100 g

This ready-to-use dyeing composition was applied to locks of natural grey hair which is 90% white for 40 minutes at 30°C. The hair was then rinsed, washed with a standard shampoo and then dried.

Locks of hair with bluish grey colour were obtained.

In this example, 1.8 g of laccase obtained from Rhus vernicifera containing 180 units/mg can be replaced by 1 g of laccase obtained from Pyricularia Orizae containing 100 units/mg sold by the company I.C.N.

EXAMPLE 2: **Dyeing composition**

The following ready-to-use dyeing composition was prepared (contents in grams):

- Laccase obtained from Rhus vernicifera containing 180 units/mg units/mg, marketed by the company SIGMA 1.8 g
- (C_8-C_{10})Alkyl polyglucoside in aqueous solution containing 60% of active substance (AS) sold under the name ORAMIX CG110 by the company SEPPIC 8.0 g
- Paraphenylenediamine 0.254 g

- 2,4-Diaminophenoxyethanol dihydrochloride 0.260 g
- Ethanol 20.0 g
- Citrulline qs pH 8.0
- Demineralized water qs 100 g

This ready-to-use dyeing composition was applied to locks of natural grey hair which is 90% white for 40 minutes at 30°C. The hair was then rinsed, washed with a standard shampoo and then dried.

Locks of hair with bluish grey colour were obtained.

In this example, 1.8 g of laccase obtained from Rhus vernicifera containing 180 units/mg can be replaced by 1 g of laccase obtained from Pyricularia Orizae containing 100 units/mg sold by the company I.C.N.

CLAIMS

1. Ready-to-use composition for the oxidation dyeing of keratinous fibres, in particular human keratinous fibres and more particularly human hair, comprising, in a carrier appropriate for dyeing keratinous fibres:

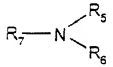
-(a) at least one enzyme of the laccase type;

-(b) at least one alkalinizing agent chosen from the group consisting of:

(i) a basic amino acid;

(ii) a compound of the following formula (A): $X(OH)_n$ in which X represents K, Li when n=1; X represents Mg, Ca when n=2; X represents $N^+R_1R_2R_3R_4$ with R_1, R_2, R_3, R_4, which are identical or different, denoting a C_1-C_4 alkyl radical, a C_1-C_4 monohydroxyalkyl or C_2-C_4 polyhydroxyalkyl radical, when n=1;

(iii) a compound of the following formula (B):



in which R_5 denotes a C_1-C_6 alkyl radical, a C_1-C_6 monohydroxyalkyl or C_2-C_6 polyhydroxyalkyl radical; R_6, R_7, which are identical or different, denote a hydrogen atom, a C_1-C_6 alkyl radical, a C_1-C_6 monohydroxyalkyl or C_2-C_6 polyhydroxyalkyl radical;

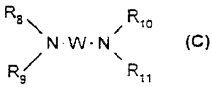
with the proviso that

• R_5, R_6, R_7 do not simultaneously denote the C_2 β-hydroxyalkyl radical,

• if R_6 and R_7 simultaneously denote H, then R_5 does not denote a C_2 monohydroxyalkyl or branched C_4 monohydroxyalkyl radical,

• if R_5 denotes hydrogen or a C_1-C_6 alkyl radical and at the same time R_6 denotes a C_1-C_6 alkyl radical, then R_7 does not denote H or a C_1-C_6 alkyl radical;

(iv) a compound of the following formula (C):



(C)

in which W is a propylene residue optionally substituted with a hydroxyl group or a C_1-C_4 alkyl radical; R_8, R_9, R_{10} and R_{11}, which are identical or different, represent a hydrogen atom, a C_1-C_4 alkyl or C_1-C_4 hydroxyalkyl radical;

- (c) at least one oxidation dye with the exception of autooxidizable indole dyes.

2. Composition according to Claim 1, characterized in that the laccase(s) are chosen from laccases of plant origin, animal origin, fungal origin, bacterial origin or are obtained by biotechnology.

3. Composition according to either of Claims 1 to 2, where the laccases are chosen from those produced by plants performing chlorophyll synthesis.

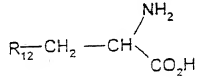
4. Composition according to Claim 3, where the laccases are chosen from those extracted from Anacardiaceae or Podocarpaceae, Rosmarinus off., Solanum tuberosum, Iris sp., Coffea sp., Daucus

carrota, Vinca minor, Persea americana, Catharenthus roseus, Musa sp., Malus pumila, Gingko biloba, Monotropa hypopithys (Indian pipe), Aesculus sp., Acer pseudoplatanus, Prunus persica, Pistacia palaestina.

5. Composition according to Claim 2, where the laccases are chosen from those derived from Pyricularia orizae, Polyporus versicolor, Rhizoctonia praticola, Rhus vernicifera, Scytalidium, Polyporus pinsitus, Myceliophtora thermophila, Rhizoctonia solani, Tramates versicolor, Fomes fomentarius, Chaetomium thermophile, Neurospora crassa, Coriolus versicol, Botrytis cinerea, Rigidoporus lignosus, Phellinus noxius, Pleurotus ostreatus, Aspergillus nidulans, Podospora anserina, Agaricus bisporus, Ganoderma lucidum, Glomerella cingulata, Lactarius piperatus, Russula delica, Heterobasidion annosum, Thelephora terrestris, Cladosporium cladosporioides, Cerrena unicolor, Coriolus hirsutus, Ceriporiopsis subvermispora, Coprinus cinereus, Panaeolus papilionaceus, Panaeolus sphinctrinus, Schizophyllum commune, Dichomitius squalens and variants thereof.

6. Composition according to any one of Claims 1 to 5, characterized in that the laccase(s) are provided in quantities ranging from 0.5 to 2000 lacu, or from 1000 to 4×10^7, or from 2×10^6 lacu units, per 100 g of composition.

7. Composition according to any one of Claims 1 to 6, characterized in that the basic amino acids correspond to the following formula (D):



(D)

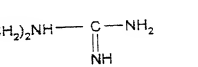
where R_{12} denotes a group chosen from:

;

$-(CH_2)_3NH_2$;

$-(CH_2)_2NH_2$

$-(CH_2)_2NHCONH_2$;



;

8. Composition according to any one of Claims 1 to 6, characterized in that the compounds of formula (B) are chosen from diethanolamine, monoisopropanolamine, diisopropanolamine, triisopropanolamine, 2-amino-2-methyl-1,3-propanediol, 2-amino-2-ethyl-1,3-propanediol, 2-amino-1-n-butanol, 1-diethylamino-2,3-propanediol, tris(hydroxymethyl)aminomethane, ethylmonoethanolamine.

9. Composition according to any one of the preceding claims, characterized in that the alkalinizing agents are used in contents by weight

ranging from 0.001% to 20%, preferably from 0.01% to 5% and still more preferably from 0.05% to 3%, relative to the total weight of the composition.

10. Composition according to any one of the preceding claims, characterized in that the oxidation dyes are oxidation bases chosen from ortho- or para-phenylenediamines, bisphenylalkylenediamines, ortho- or para-aminophenols, and heterocyclic bases, as well as the addition salts of these compounds with an acid.

11. Composition according to Claim 10, characterized in that the oxidation bases are present in concentrations ranging from 0.0005 to 12% by weight relative to the total weight of the composition.

12. Composition according to Claim 10, characterized in that the oxidation dyes are couplers chosen from meta-phenylenediamines, meta-aminophenols, meta-diphenols, heterocyclic couplers, and the addition salts of these compounds with an acid.

13. Composition according to Claim 12, characterized in that the couplers are present in concentrations ranging from 0.0001 to 10% by weight relative to the total weight of the composition.

14. Composition according to any one of Claims 10 to 13, characterized in that the addition salts with an acid of the oxidation dyes are chosen from hydrochlorides, hydrobromides, sulphates, tartrates, lactates and acetates.

15. Composition according to any one of Claims 1 to 14, characterized in that it contains, in addition, direct dyes.

16. Composition according to any one of Claims 1 to 15, characterized in that the medium appropriate for keratinous fibres (or carrier) consists of water or of a mixture of water and of at least one organic solvent.

17. Composition according to Claim 16, characterized in that the organic solvents may be present in proportions preferably ranging from 1 to 40% by weight approximately relative to the total weight of the composition, and still more preferably ranging from 5 to 30% by weight approximately.

18. Composition according to any one of Claims 1 to 17, characterized in that the pH varies from 4 to 11 approximately, and preferably from 6 to 9 approximately.

19. Composition according to any one of Claims 1 to 28, characterized in that it contains, in addition, at least one cosmetic adjuvant conventionally used in hair dyeing compositions, chosen from the group consisting of surfactants, polymers, thickeners, antioxidants, enzymes different from the laccases, penetrating agents, sequestering agents, perfumes, dispersing agents, film-forming agents, screening agents, vitamins, preservatives or opacifying agents.

20. Method of dyeing keratinous fibres, and in particular human keratinous fibres such as hair, characterized in that at least one ready-to-use dyeing composition as defined in any one of Claims 1 to 19 is applied to the said fibres for a sufficient time to develop the desired colour.

21. Method according to Claim 20, characterized in that it comprises a preliminary step consisting in storing in a separate form, on the one hand, a composition (A) comprising, in a medium appropriate for dyeing, at least one oxidation dye as defined in any one of Claims 1 and 10 to 14 and on the other hand, a composition (B) containing, in a medium appropriate for keratinous fibres, at least one enzyme of the laccase type as defined in any one of Claims 1 to 6, and then in mixing them at the time of use before applying this mixture to the keratinous fibres; the composition (A) or the composition (B) containing the alkalinizing agent as defined in Claims 1 and 7 to 9.

22. Multicompartment device or dyeing "kit", characterized in that it comprises a first compartment containing the composition (A) as defined in Claim 21 and a second compartment containing the composition (B) as defined in Claim 21.

PTO/SB/105 (8-96)
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Declaration and Power of Attorney for Patent Application

Déclaration et Pouvoir pour Demand de Brevet

French Language Declaration

En tant que l'inventeur nommé ci-après, je déclare par le présent acte que:

As a below named inventor, I hereby declare that:

Mon domicile, mon adresse postale et ma nationalité sont ceux figurant ci-dessous à côté de mon nom.

My residence, post office address and citizenship are as stated next to my name.

Je crois être le premier inventeur original et unique (si un seul nom est mentionné ci-dessous), ou l'un des premiers co-inventeurs originaux (si plusieurs noms sont mentionnés ci-dessous) de l'objet revendiqué, pour lequel une demande de brevet a été déposée concernant l'invention intitulée

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

DYEING COMPOSITION CONTAINING A LACCASE AND KERATINOUS FIBRE DYEING METHOD

et dont la description est fournie ci-joint à moins que la case suivante n'ait été cochée:

the specification of which is attached hereto unless the following box is checked:

☐ a été déposée le _____
sous le numéro de demande des Etats-Unis ou le numéro de demande international PCT
_____ et modifiée
_____ (les cas échéant).

☒ was filed on December 21, 1998 as United States Application Number or PCT International Application Number PCT/FR98/02805 and was amended on _____ (if applicable).

Je déclare par le présent acte avoir passé en revue et compris le contenu de la description ci-dessus, revendications comprises, telles que modifées par toute modification dont il aura été fait référence ci-dessus.

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above

Je reconnais devoir divulguer toute information pertinente à la brevetabilité, comme défini dans le Titre 37, § 1.56 du Code fédéral des réglementations.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56.

PTO/SB/105 (8-96)
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Attorney Docket No.: 05725.0623

French Language Declaration

Je revendique par le présent acte avoir la priorité étrangère, en vertu du Titre 35, § 119(a)-(d) ou § 365(b) du Code des Etats-Unis, sur toute demande étrangère de brevet ou certificat d'inventeur ou, en vertu du Titre 35, § 365(a) du même Code, sur toute demande internationale PCT désignant au moins un pays autre que les Etats-Unis et figurant ci-dessous et, en cochant la case, j'ai aussi indiqué ci-dessous toute demande étrangère de brevet, tout certificat d'inventeur ou toute demande internationale PCT ayant une date de dépôt précédant celle de la demande à propos de laquelle une priorité est revendiquée.

I hereby claim foreign priority under Title 35, United States Code, § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International Application which designated at least one country other than the United States, listed below, and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

Prior foreign application(s) Demande(s) de brevet antérieure(s)		Priority Not Claimed Droit de priorité non revendiqué
98/00,250 (Number) (Numéro)	France (Country) (Pays)	13 January 1998 (Day/Month/Year Filed) (Jour/Mois/Année de dépôt) ☐
(Number) (Numéro)	(Country) (Pays)	(Day/Month/Year Filed) (Jour/Mois/Année de dépôt) ☐

Je revendique par le présent acte tout bénéfice, en vertu du Titre 35, § 119(e) du Code des Etats-Unis, de toute demande de brevet provisoire effectuée aux Etats-Unis et figurant ci-dessous.

I hereby claim the benefit under Title 35, United States Code, § 119(e) of any United States provisional application(s) listed below.

(Application No.) (N° de demande)	(Filing Date) (Date de dépôt)
(Application No.) (N° de demande)	(Filing Date) (Date de dépôt)

Je revendique par le présent acte tout bénéfice, en vertu du Titre 35, § 120 du Code des Etats-Unis, de toute demande de brevet effectuée aux Etats-Unis, ou en vertu du Titre 35, § 365(c) du même Code, de toute demande internationale PCT désignant les Etats-Unis et figurant ci-dessous et, dans la mesure où l'objet de chacune des revendications de cette demande de brevet n'est pas divulgué dans la demande antérieure américaine ou internationale PCT, en vertu des dispositions du premier paragraphe du Titre 35, § 112 du Code des Etats-Unis, je reconnais devoir divulguer toute information pertinente à la brevetabilité, comme défini dans le Titre 37, § 1.56 du Code fédéral des réglementations, dont laquelle est devenue disponible entre la date de dépôt de la demande antérieure, et la date de dépôt de la demande nationale ou internationale PCT de la présente demande:

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s), or § 365(c) of any PCT International Application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International Application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose any or all information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

(Application No.) (N° de demande)	(Filing Date) (Date de dépôt)	(Status) (patented, pending, abandoned) (Status) (breveté, en cours d'examen, abandonné)
(Application No.) (N° de demande)	(Filing Date) (Date de dépôt)	(Status) (patented, pending, abandoned) (Status) (breveté, en cours d'examen, abandonné)

Je déclare par le présent acte que toute déclaration ci-incluse est, à ma connaissance, véridique et que toute déclaration formulée à partir de renseignements ou de suppositions est tenue pour véridique; et de plus, que toutes ces déclarations ont été formulées en sachant que toute fausse déclaration volontaire ou son équivalent est passible d'une amende ou d'une incarcération, ou des deux, en vertu de la Section 1001 du Titre 18 du Code des Etats-Unis, et que de telles déclarations volontairement fausses risquent de compromettre la validité de la demande de brevet ou du brevet délivré à partir de celle-ci.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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French Language Declaration

POUVOIRS: En tant que l'inventeur cité, je désigne par la présente l'(les) avocat(s) et/ou agent(s) suivant(s) pour qu'ils poursuive(nt) la procédure de cette demande de brevet et traite(nt) toute affaire s'y rapportant avec L'Office des brevets et des marques: *(mentionner le nom et le numéro d'enregistrement).*

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